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RAMAN AND PHONON MASERS^g

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RAMAN AND PHONON MASERS

When the very intense beams of light generated by optical masers interact with matter, they tend to excite various degrees of freedom of both the matter and the radiation field, and sometimes such excitations are quite large. This must be expected because the effective temperature of maser beams is in the range 10^{15} to 10^{30} °K, so that any interaction with cooler matter must tend to degrade the beam energy, and any equilibrium between the light beam and a small number of degrees of freedom must still correspond to enormously high temperatures. At certain threshold intensities, instabilities set in which transfer energy very rapidly from the beam to other electromagnetic or mechanical modes of the system. Some of these processes will be discussed here, in particular the generation of coherent molecular oscillations or phonons by interaction between intense maser beams and matter. Such instabilities produce the observed intense Raman radiation along with coherent molecular vibrations. They should also lead to generation of intense high-frequency sound waves and of infrared radiation.

Stimulated Raman Effects, Coherent Molecular
Vibrations, and Light Modulation

Normal Raman scattering, which has been known for some¹time,
corresponds to spontaneous emission of Raman radiation. It can be
described in terms of individual molecules and photons in which such
Raman emission from one molecule is incoherent with that from a neighboring
one. The process shows features of a distinctly quantum-mechanical nature.

A Raman maser, or stimulated Raman emission, typically involves
large numbers of molecules and photons. Except for details of molecular
characteristics or initiation of the radiation by spontaneous emission,
these processes may be discussed rather satisfactorily from a classical
point of view.

The potential energy of a molecule with polarizability α in an
electric field $\vec{E}(\vec{r}, t)$ is

$$W = - \frac{1}{2} \alpha(x) \left| \vec{E}(\vec{r}, t) \right|^2 .$$

The dependence of α on x , a molecular vibrational or rotational coordinate,
molecule to the electro-
results in non-linear coupling of the magnetic field and hence in scattered

Raman radiation of shifted frequency. As an example we might consider a diatomic molecule for which x describes the deviation from the equilibrium internuclear distance during vibration; $\alpha = \alpha_0 + \alpha_1 x$ in the lowest order approximation. The frequency dependence and tensorial properties of α will be neglected. The force driving such vibrations will be

$$-\frac{\partial W}{\partial x} = \frac{1}{2} \alpha_1 E^2$$

A sinusoidally varying force, $F \cos \omega t$, will give the following differential equation for the vibrational coordinate:

$$m \ddot{x} + R \dot{x} + f x = F \cos \omega t.$$

R is a phenomenological damping coefficient given by the half-width $\Delta\nu$ of the resonance curve:

$$\Delta\nu = R/2\pi m.$$

It may be noted that in this approximation the molecular oscillator itself is, in the absence of an electromagnetic field, completely governed by a linear equation. The dielectric response of the molecules will also be assumed completely linear in the absence of molecular vibration. It

is the coupling between the two motions which introduces a non-linearity.

The solution for x at frequencies near the resonance frequency $\omega_r = \sqrt{f/m}$

is $x = (F/R\omega) \sin \omega t$. Thus the molecular vibration is 90° out of phase with the driving force.

The ratio of the dynamic stretch of the molecule near resonance to the static stretch for equal driving forces is $x_{\text{dyn.}} / x_{\text{stat.}}$

$= f/R\omega = \frac{\nu_r}{\Delta\nu}$. In liquids this ratio is frequently as large as 10^3 . The amplitude of vibration can thus become quite high, and non-linear coupling effects associated with the vibration will show up much more prominently at this resonance than at nearby frequencies. Thus, the treatment below will consider only the resonant situation.

The generation of stimulated Raman emission requires the radiation field to contain radiation of Raman-shifted frequency as well as unshifted light. Thus consider E_0 , the electric field of the initial coherent radiation at frequency ω_0 ; and E' , radiation of frequency $\omega' = \omega_0 \pm \omega_r$, initiated by spontaneous Raman emission. ω_r is the resonant vibrational frequency of the molecule. For traveling waves the total electric field

is then $\vec{E} = \vec{E}_0 \cos (\omega_0 t - \vec{k}_0 \cdot \vec{r}) + \vec{E}' \cos (\omega' t - \vec{k}' \cdot \vec{r} + \phi')$.

The resonant vibration of the molecules produced by the beating of these two light waves through the force $\frac{1}{2} \alpha_1 E^2$ is, assuming E is coherent over times much larger than the inverse line width $1/\Delta\nu$,

$$x = \frac{E_0 E' \alpha_1}{2 R (\omega_0 - \omega')} \sin \left[(\omega_0 - \omega') t - (\vec{k}_0 - \vec{k}') \cdot \vec{r} - \phi' \right].$$

The electric dipole moment per unit volume induced by the field \vec{E} is

$\vec{\mu} = \alpha \vec{E}$. This oscillating dipole moment may interact with the electric field $\vec{E}' \cos (\omega' t - \vec{k}' \cdot \vec{r} + \phi')$ to increase or decrease its amplitude continually, as long as the phase ϕ' is approximately constant. The time-averaged rate of energy flow per unit volume to the field \vec{E}' is

$$N \left\langle \frac{d\vec{\mu}}{dt} \cdot \vec{E}' \right\rangle. \text{ Here } N \text{ is the effective number of molecules}$$

per unit volume which take part in this interaction; ~~from this follows~~ ^{or}

$N_0 - N_1$ where N_0 is the density of molecules in the ground state involved

and N_1 , the density in the upper state. Hence the power per unit volume

delivered to E' from the input beam E_0 is:

$$P' = \frac{N \alpha_1^2}{8 R} \frac{\omega'}{\omega_0 - \omega'} (\vec{E}_0 \cdot \vec{E}')^2.$$

For Stokes radiation in which $\omega' = \omega_0 - \omega_r$, $P' > 0$ so that any radiation E' is amplified. The phase φ' is arbitrary as long as it does not vary much during the molecular relaxation time $\frac{1}{2\pi\Delta\nu}$.

For anti-Stokes radiation $\omega' = \omega_0 + \omega_r$, $P' < 0$ and any initial radiation E' will lose energy*.

Since P' is quadratic in E' , there will be a threshold intensity required for E_0 before the instability takes place, allowing E' to build up. For build-up of the traveling wave $\vec{E'}$, the fractional ^{power} gain per unit

length $a = \frac{N \pi}{c R} \alpha_1^2 \frac{\omega_0 - \omega_r}{\omega_r} E_0^2$, must be greater than or equal to the fractional loss per unit length b ^{which may, for example, be due to absorption.}. This leads to

the requirement:

$$E_0^2 > \frac{b c R}{\pi N \alpha_1^2} \frac{\omega_r}{\omega_0 - \omega_r}.$$

Once this threshold condition on intensity of the "pump" field E_0 is met, coherent Stokes radiation will increase from the spontaneous Raman emission with the exponential buildup usual in maser theory.

* Anti-Stokes radiation would be amplified and Stokes radiation de-amplified if there were an inversion of population in the Raman material due to an additional excitation source, since then $N_0 - N_1$ would be negative.

One might also ask whether there will always be an initial wave \vec{E}' generated by spontaneous Raman emission. Usually, the spontaneous Raman emission will be sufficiently rapid to easily supply the initial field which is amplified. Consider a "plane" wave, defined in angular spread by necessary diffraction due to the finite dimensions of the space in which it exists. The time required for this plane wave to be doubled by stimulated emission is just equal to the average time required for a quantum to be emitted into it by spontaneous emission. The probability of spontaneous emission in any other "plane" wave of similar solid angle is identical, so that if a larger solid angle of waves can be amplified the effective spontaneous emission rate for initiation of the wave E' is correspondingly greater.

Gain in a Raman maser has been formulated several times previously,^{2,3,4} and Zeiger and Tannenwald⁴ have noted that coherent molecular oscillations must exist in such a maser. Usually, the result is expressed in terms of a sum of matrix elements, which are separately very difficult to evaluate. In the above expression, these are replaced by the single quantity α_i .

R may be evaluated from the half-width $\Delta\nu$ of the Raman resonance; for a diatomic molecule $R = 2\pi m \Delta\nu$, where m is the reduced mass. Here homogeneous broadening has been assumed, but extension to the inhomogeneous case is straightforward and gives the same final result when expressed in terms of the half-width $\Delta\nu$ of the Raman line. In typical cases the gain a due to stimulated emission in a pumping beam of power 100 megawatts per square centimeter is of the order of 10 cm^{-1} . If the absorption b is about 10% per cm., this means that the threshold would be at about one megawatt/cm² of pump power. As is usual in masers, high gain results in an emission line which is appreciably narrower than that for spontaneously emitted Raman radiation.

For a ruby pulse with the order of 100 M watts/cm² in a single mode, molecules are stretched by about 10^{-4} of the bond distance. In planes defined by $(\vec{k}_0 - \vec{k}_{-1}) \cdot \vec{r} = \text{constant}$, vibrations of all molecules are in phase, and hence the molecules push each other apart. The resulting expansion in the planes $(\vec{k}_0 - \vec{k}_{-1}) \cdot \vec{r} = \text{constant}$ cannot, however, occur as rapidly as the molecular oscillation, since this would imply macroscopic motions of enormous kinetic energy. The gross expansion of the material

occurs, instead, in a time as large as about 10^{-9} sec., and is determined by the average value of E^2 over intervals of times as long as this. The resulting spatial variations in dielectric susceptibility are of the order 10^{-4} . In addition, there are variations in the susceptibility of the same order which oscillate at the molecular frequency, associated with the variation α_1 of molecular polarizability with molecular stretching. There is thus a slowly varying susceptibility and one varying synchronously with the molecular vibration, each periodic in $\vec{k}_0 - \vec{k}_{-1}$. These variations act like a three-dimensional phase grating and scatter light waves; those varying at molecular vibrational frequencies not only scatter, but also modulate light waves.

To be able to understand in detail the generation of anti-Stokes radiation, we consider the case where electromagnetic waves of both Stokes and anti-Stokes frequencies are initially present,

$$\begin{aligned} \vec{E} = & \vec{E}_0 \cos (\omega_0 t - \vec{k}_0 \cdot \vec{r}) + \vec{E}_{-1} \cos [(\omega_0 - \omega_r) t - \vec{k}_{-1} \cdot \vec{r} + \varphi_{-1}] \\ & + \vec{E}_1 \cos [(\omega_0 + \omega_r) t - \vec{k}_1 \cdot \vec{r} + \varphi_1]. \end{aligned}$$

As in the treatment above⁵, the vibration of the molecule and its oscillating

dipole moment are calculated. The average power gain per unit volume of

the Stokes radiation is:

$$P_{-1} = \frac{N\alpha_1^2}{8R} \frac{\omega_0 - \omega_r}{\omega_r} \left\{ (\vec{E}_0 \cdot \vec{E}_{-1})^2 + (\vec{E}_0 \cdot \vec{E}_1) (\vec{E}_0 \cdot \vec{E}_{-1}) \cos \left[(2\vec{k}_0 - \vec{k}_1 - \vec{k}_{-1}) \cdot \vec{r} + \varphi_1 + \varphi_{-1} \right] \right\},$$

and of the anti-Stokes wave:

$$P_{+1} = \frac{N\alpha_1^2}{8R} \frac{\omega_0 + \omega_r}{\omega_r} \left\{ - (\vec{E}_0 \cdot \vec{E}_1)^2 - (\vec{E}_0 \cdot \vec{E}_1) (\vec{E}_0 \cdot \vec{E}_{-1}) (2\vec{k}_0 - \vec{k}_1 - \vec{k}_{-1}) \cdot \vec{r} + \varphi_1 + \varphi_{-1} \right\}.$$

From these expressions it is evident that there is a solution for power gain at the Stokes frequency when anti-Stokes radiation is absent, which is just that discussed above. However, there is no possibility of gain in the anti-Stokes line unless the Stokes radiation is present. Thus there is a phase independent term which always gives gain for Stokes and loss for anti-Stokes. It is hence expected that $\vec{E}_{-1} > \vec{E}_1$, and when this is true there may be gain for anti-Stokes radiation if $2\vec{k}_0 = \vec{k}_{-1} + \vec{k}_1$ and the phase $\varphi_1 + \varphi_{-1}$ is suitable, $\varphi_1 + \varphi_{-1} = \pi$ giving the maximum gain.

If the phase $\varphi_1 + \varphi_{-1}$ is such that gain can occur for anti-Stokes

radiation, the gain for the Stokes radiation is then always decreased at resonance by the presence of anti-Stokes waves. In this case the amplitude of the molecular vibration is proportional to $\vec{E}_{-1} - \vec{E}_1$; that is, the two fields drive the molecule in opposite phase.

The generation of the anti-Stokes line E_1 may be interpreted as caused by a modulation of the input radiation E_0 due to the variations of index of refraction at frequency ω_1 which result from the coherent molecular vibrations set up by E_0 and the existing E_{-1} . Such modulation produces sidebands on any radiation present, and hence if the threshold condition on E_0 for the generation of E_{-1} is met, many frequencies can be produced without a further threshold.

Since there is no condition on the wave vectors from the gain term for Stokes radiation, it will be radiated into all directions although under ideal conditions the gain would have an angular dependence $\cos^2 \theta$, where θ is the angle between polarization of the pump and Stokes radiation. On the other hand, the wave vector relation, $2\vec{k}_0 = \vec{k}_1 + \vec{k}_{-1}$ required for gain

in the anti-Stokes line confines this latter radiation to a cone in the forward direction, with half-angle θ_1 , around the initial beam. This vector relation has been also given by Terhune,⁶ although apparently on the basis of a 4-quantum interaction rather than the mechanism discussed here. ~~It is a rather general relation which would apply to almost any~~

~~proposed mechanism for generation of E_1 and E_2 .~~ For small angles one can

show that the above relation gives

$$\theta_1^2 \approx \frac{1}{n} \frac{\omega_0 - \omega_r}{\omega_0 + \omega_r} \left[(\Delta n_1 - \Delta n_{-1}) + \frac{\omega_r}{\omega_0} (\Delta n_1 + \Delta n_{-1}) \right].$$

Here $n = n(\omega_0)$ is the index of refraction at frequency ω_0 ;

$$\Delta n_1 = n(\omega_0 + \omega_r) - n(\omega_r), \text{ and } \Delta n_{-1} = n(\omega_0) - n(\omega_0 - \omega_r).$$

Usually the first term in the bracket of this expression predominates, so

that the angle θ_1 is determined by the curvature of the dispersion curve.

The Stokes radiation which interacts with the anti-Stokes wave occurs at an

$$\text{angle: } \theta_{-1} = \frac{\omega_0 + \omega_r}{\omega_0 - \omega_r} \theta_1.$$

These angles are typically of the order of a few degrees. As was stated

above, there is no additional threshold needed for the generation of E_1

beyond the existence of E_{-1} at the appropriate angle, since the gain is not quadratic in E_1 .

by the above mechanism
Anti-Stokes radiation will not usually build up/in a Raman maser with plane parallel reflectors perpendicular to the initial beam. Stokes radiation will build up by the first process discussed above in the direction of maximum gain which is parallel to the pump beam, at the expense of possible Stokes waves in other directions. Because of dispersion, the above wave vector relation required for the generation of anti-Stokes radiation cannot be satisfied by parallel E_0 and E_{-1} waves.

A field E_{-2} at frequency $\omega_0 - 2\omega_r$ can be emitted diffusely by a process generating power proportional to $E_1^2 E_{-2}^2$ essentially identical with that for the generation of E_{-1} . In addition, it may be produced through modulation of E_{-1} by the oscillations in dielectric constant due to E_0 and E_{-1} , giving a power generation proportional to $E_0 (E_{-1})^2 E_{-2}$. The latter case, which has no threshold provided E_{-1} is present, is the more important in generation of Raman radiation by intense beams outside a cavity, since in any one direction $|E_{-2}| < |E_0|$. This process requires

$\vec{k}_0 - \vec{k}_{-1} = \vec{k}_{-1} - \vec{k}_{-2}$ where the wave vectors \vec{k}_{-1} and \vec{k}_{-2} may be differently oriented but both correspond to frequencies $\omega_0 - \omega_r$. This equation cannot usually be satisfied in a dispersive medium if E_{-1} is in the same direction as E_0 . Hence the former mechanism, which has a threshold, is probably the more important in a resonant cavity. Similar mechanisms can generate Stokes radiation of frequencies $\omega_0 - n\omega_r$. The strongest such radiation will usually be due to the modulation processes, which require no threshold condition, and will be diffusely emitted unless there is some feedback by reflection of the wave.

Radiation of frequency $\omega_0 + 2\omega_r$ is produced without threshold effects by vibrational modulation of $\omega_0 + \omega_r$ and is emitted in directions specified by $\vec{k}_0 - \vec{k}_{-1} = \vec{k}_2 - \vec{k}_1$. For normal materials, there is a \vec{k}_{-1} and a \vec{k}_2 which will satisfy this equation, the angle between \vec{k}_0 and \vec{k}_2 being of the order of $2\theta_1$. Other anti-Stokes beams of frequency $\omega_0 + n\omega_r$ are similarly generated in cones about the original beam.

Although the buildup of Stokes radiation is exponential in the absence of anti-Stokes radiation, the interaction of the two fields produces

a linear gain. If the fields E_{-1} and E_1 are initially $E_{-1}(0)$ and 0 respectively, and if each has a fractional power loss b per unit length in the medium due to other effects, the build-up thereafter of the two interacting waves in a distance L will have the form

$$E_1 = E_{-1}(0) \frac{aL}{2} e^{-\frac{bL}{2}}$$

$$E_{-1} = E_{-1}(0) \left(\frac{aL}{2} + 1 \right) e^{-\frac{bL}{2}}$$

Thus for $aL/2 \gg 1$, E_1 becomes comparable to E_{-1} . In this case, $E_{-1}(0)$ is understood to be not noise, as is often the case in masers, but Stokes light of sizeable intensity, perhaps scattered from other directions in which this radiation experiences an exponential buildup. If $E_{-1}(0)$ is large, the buildup of E_1 and E_{-1} in the directions specified by the wave vector equation may be considerable. Usually $bL/2 \ll 1$ for pertinent cases.

Infrared Generation

The molecules set into vibration in the crystal are all vibrating coherently over some spatial distribution. If these molecules have dipole moments, then their vibrating dipoles can coherently radiate infrared

radiation at the Raman frequency providing that the wave vector relation $\vec{k}_0 - \vec{k}_{-1} = \vec{k}_r$ is satisfied. Here \vec{k}_r is the wave vector of the infrared radiation. From this relation, one finds that the angle θ_r between the generated infrared beam and the incident radiation is given by

$$\cos \theta_r = 1 + \frac{\Delta n_r}{n} \frac{\omega_0 - \omega_r}{\omega_0} + \frac{\Delta n_{-1}}{n} \frac{(\omega_0 - \omega_r)^2}{\omega_0 \omega_r}$$

Here Δn_r is $n(\omega_0) - n(\omega_r)$. If both Δn_r and Δn_{-1} are positive,

θ_r is no real angle, since $\cos \theta_r > 1$. However, Δn_r will be negative for many substances if ω_r is an infrared frequency and Δn_{-1} may on occasion be negative, so that θ_r can be real. When this condition is not met, infrared radiation can still be radiated from surface layers of material of finite extent.

Consider now the origin of the dipole moment which is oscillating.

Normally, the Raman transition is not thought of as involving an oscillating dipole moment, but this only holds when the molecules have a center of inversion symmetry. If they do not have such a center, then the transition can be both infrared and Raman active. If a center of symmetry normally exists, it can be removed by applying a strong d.c. field which will then allow both infrared and Raman activity in the molecules.

One might wonder whether the infrared radiation will be strongly re-absorbed in traversing the material, since it corresponds to an active molecular transition. It will not be absorbed, but amplified, if it has the correct phase, since all the molecules are oscillating coherently. A wave with an arbitrary phase may be absorbed, but with just the correct phase it will receive energy from each molecule in turn as it propagates, as a result of the matching of wave vectors. The usual absorption then becomes an emission.

This is another example of amplification where there is no inversion of population. The numerical probability of finding a molecule in its upper state is typically only about 10^{-6} , corresponding to one part in 10^3 of the upper state wave function mixed with the lower-state function. Amplification occurs not because of a population inversion, but because of phase coherence between the various oscillating systems, so that an infrared wave of the correct phase and wave vector can receive energy from every oscillator.

Phonon Masers

The phonon field is very similar in structure to the electromagnetic field, and hence one may expect maser-like phenomena in acoustic waves. The theory of a phonon maser quite analogous to a solid-state maser in the microwave region was discussed some time ago^{7,8} and has been realized by Tucker.⁹ He used impurity atoms in a crystalline lattice and inverted their population. Since the excited atoms in the lattice interact with the phonon field--as well as with the electromagnetic field--they can provide a phonon maser if the usual type of critical condition for amplification or oscillation is met.

Here a different type of phonon generation will be discussed, namely, one based on stimulated Brillouin scattering. In the normal Brillouin scattering,¹⁰ an incident light beam is scattered by thermally excited acoustic waves. The scattered light is shifted in frequency and the energy difference taken up by the acoustic wave just as in the usual Raman effect. However, because the scattering is produced by a large region of the crystal rather than by a localized molecule, as in the usual Raman effect, the scattered beam is highly directional. If the incident

beam is sufficiently intense, this process can be made to produce coherent stimulated emission of both the scattered light and the acoustic wave.

The process may be viewed as analogous to a Raman maser, or to parametric amplification of an extended wave.

The coupling of the light waves with the sound waves which will be considered is due to electrostriction, the compression or expansion of the material by electric fields in the light wave. For a quantitative measure of this effect, consider a condenser of volume V filled with a compressible fluid. The change of electrical energy when the fluid contracts by an amount ΔV is $\frac{E^2}{8\pi} V \Delta\epsilon$ where $\Delta\epsilon = \frac{d\epsilon}{dV} \Delta V$ is the change in dielectric constant due to compression. This equals the work done on the fluid, $p\Delta V$, where p is the electrostrictive pressure. Since $\frac{\Delta p}{p} = - \frac{\Delta V}{V}$, this yields $p = \frac{E^2}{8\pi} \rho \frac{d\epsilon}{dp} = \frac{\chi E^2}{8\pi}$ where $\chi = \rho \frac{d\epsilon}{dp}$. Usually $\rho \frac{d\epsilon}{dp}$ is of order unity for liquids or solids, so that for a power flux of 300 M Watts/cm² the electrostrictive pressure p from this expression is about 10^5 dyne/cm².

Now consider two beams of light at slightly different frequencies in the medium. Since the electrostatic pressure is proportional to E^2 , the medium will be compressed at the beat frequency of the two beams.

(which may be anything up to the frequency of the light). So under certain conditions acoustic waves of high energy will build up. The whole crystal can be considered as one molecule and to vibrate analogously to a single molecule in the Raman effect discussed above. We are, in fact, considering a Raman effect of gross material. The crystal vibrates in modes, the wavelengths of which are characteristically short compared to the dimensions of the crystal. Since the susceptibility depends on the compression, the acoustic modes represent spatial phase gratings. The theory of the interaction between light waves and the acoustic modes is similar to the above treatment of light waves interacting with vibrating molecules. Its quantitative behavior may be derived as follows.

The change in polarization per unit volume due to a pressure p in an isotropic material is given by $\vec{\mu} = \frac{\vec{E}}{4\pi} \frac{d\epsilon}{dp} \frac{dp}{dp} \quad p = \frac{1}{4\pi} \frac{\chi}{B} \vec{E} p.$

Hence, a non-linear coupling between the electromagnetic field and the compressional waves is produced by the fluctuating dipole moment. Here B is the bulk modulus, or the inverse of the adiabatic compressibility, $\frac{1}{B} = \frac{1}{\rho} \frac{d\rho}{dp}.$

Only a general outline of the theory of build-up of coupled acoustic and electromagnetic waves will be presented. The polarization produced by

the acoustic wave feeds energy into the electromagnetic wave. The power gain per unit volume of the electromagnetic wave is:

$$(\text{Power}) \text{ e.m.} = -\vec{E} \cdot \frac{d\vec{\mu}}{dt} = -\frac{1}{4\pi} \frac{\chi}{B} E \frac{d}{dt} (E p).$$

The power fed into the sonic wave per unit volume is

$$(\text{Power}) \text{ sonic} = -p \oint \frac{dv}{dt} = -p \oint \frac{dv}{dp} \frac{dp}{dt} = +\frac{p}{B} \frac{dp}{dt}.$$

From the formula for the electrostrictive pressure we obtain:

$$(\text{Power}) \text{ sonic} = +\frac{1}{4\pi} \frac{\chi}{B} p \frac{dE^2}{dt}.$$

Consider three electromagnetic waves of different frequencies present in the electrostrictive medium:

$$\begin{aligned} \vec{E} = & \vec{E}_0 \cos(\omega_0 t - \vec{k}_0 \cdot \vec{r}) + \vec{E}_{-1} \cos(\omega_{-1} t - \vec{k}_{-1} \cdot \vec{r} + \varphi_{-1}) \\ & + \vec{E}_1 \cos(\omega_1 t - \vec{k}_1 \cdot \vec{r} + \varphi_1) \end{aligned}$$

and a sonic wave:

$$p_s = p \cos(\omega_s t - \vec{k}_s \cdot \vec{r} + \varphi_s)$$

$$\text{Here } \omega_{-1} = \omega_0 - \omega_s \text{ and } \omega_1 = \omega_0 + \omega_s.$$

The power fed into each one of these waves may now be calculated. It is

easy to show that, as in stimulated Raman effects discussed above, waves of

the upconverted frequency ω_1 is not amplified. But the E_{-1} wave and

the sonic wave may be amplified. Thus one need consider only three interacting waves in the system.

Amplification will take place only when the phase matching condition $\vec{k}_0 = \vec{k}_1 + \vec{k}_s$ is fulfilled. From this condition for the wave vectors the frequency of the sonic wave is found to be $\omega_s = 2 \frac{v_s}{c'} \omega_0 \sin \frac{\theta}{2}$ where v_s is the velocity of sound in the medium, c' is the velocity of light, and (θ) is the angle between incident and scattered light waves with frequency difference ω_s . This expression is a valid approximation when $v_s \ll c'$. It was first derived by Brillouin.¹⁰

The velocity ratio $\frac{v_s}{c'}$ is such that the sonic frequencies can go up to about 10^{11} cps or even 10^{12} cps for material with a very high velocity of sound. Thus the frequencies lie in the microwave frequency region. By calculating the power gain per unit length the threshold condition is found

$$\frac{E_0^2}{8\pi} \geq \frac{\epsilon_0 B}{\gamma^2} \frac{1}{k_s L_s k_1 L_1} .$$

Here L_1, L_s are the absorption lengths of light and sound waves respectively.

When the input radiation fulfills this condition, the sonic waves and the

scattered electromagnetic wave field begin to build up. For absorption lengths $L_1 \simeq 10^2$ and $L_s \simeq 10^{-2}$ (e.g. quartz) and normal bulk moduli the necessary input power to meet this threshold is found to be about 1 Megawatt/cm².

The maximum power which can go into the sound waves is ultimately governed by the Manley-Rowe relations, that is, by the ratio of the optical to the sonic frequencies. Thus with 1 M Watt of power in the light beam a maximum of there can be [^]about 1 K Watt of power in the sonic waves. It is particularly interesting that by this method acoustic energy can be fed into the interior of nearly every optical material. By the present known techniques, high frequency acoustic power may be generated in quartz crystals, but cannot easily be coupled into many other materials.

More General Treatment of Sonic Waves

So far we have treated the system in rather an elementary way. We ^{there are} have assumed that ~~we have~~ sonic and electromagnetic waves existing in the medium and have tried to find out under what conditions power is fed into them from the initial electromagnetic radiation or "pumping" wave. This

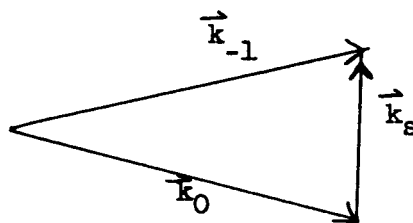
approach is good in many cases, but it has its limitations. The fields E and p have been assumed to exist in the same space and to continue so.

This is perfectly valid in a crystal when the fields E and p are standing waves reflected back and forth, that is, for a resonant type of structure.

For two traveling waves, such an assumption is not adequate unless the sonic and electromagnetic waves are traveling in the same direction, in which case they tend to remain together and the above equations can be

valid. However, the condition $\vec{k}_0 = \vec{k}_{-1} + \vec{k}_s$ requires that usually the sonic wave and scattered light waves travel in opposite directions. This is because $\omega_s \ll \omega$, even though k_s and k_0 may be comparable in magnitude.

The vectors \vec{k}_0 and \vec{k}_{-1} are therefore approximately equal in length, though usually different in direction. Hence the vector diagram



has nearly the form of an isosceles triangle and the sound wave propagates at an angle greater than 90° with respect to the scattered light wave. If

there is gain, the light wave and sound wave therefore build up in different **traveling wave**

directions, and hence one must treat this [^]case more carefully than we have

done so far.

A ^{more}~~proper~~ general approach is to start with the wave equations:

$$\ddot{\rho}_s + \alpha_s \dot{\rho}_s - v_s^2 \nabla^2 \rho = - \frac{\gamma}{c_a} \nabla^2 E^2$$

$$\ddot{E} + \alpha \dot{E} - c^2 \nabla^2 E = - \frac{\gamma}{\epsilon_0} \frac{\partial^2}{\partial t^2} (\rho E) .$$

The left-hand sides of each of these equations are the normal wave equations

with damping constants α_s and α . The right-hand sides represent non-

linear driving or coupling terms. These equations have been given by Kroll,¹¹

who has also obtained certain classes of solutions for them. They must be

solved subject to boundary conditions appropriate to the system of interest.

Details of this method are very similar to those in Kroll's discussion of

parametric oscillation.¹² This approach allows, of course, appropriate

discussion of either resonant or traveling-wave systems.

Uses of Such Effects

How fast does the acoustic wave build up in the crystal? For light pulses as short as 10^{-8} sec. the wave may not build up fast enough to be

very important. For example, if one has an electric field strength just twice that required to overcome the threshold condition, then the rate of buildup of the acoustic wave is about equal to the rate of loss of the least lossy wave; this is usually that of the light wave, and represents a rather slow buildup. However, one can exceed the threshold field by factors of the order of 100 fairly easily and so obtain conditions in which the sound wave builds up importantly within the duration of a typical pulse.

A strong sound-wave field can also result if in the initial beam both of the fields E_0 and E_{-1} are present. This case is not uncommon, for usually one has the frequencies of several modes in the beam. We therefore start off with a pressure wave which gives an acoustic shock to the crystal as a result of the electrostriction. Some of the frequencies in the shock can be resonant in the system and so build up, or they may simply build up as traveling waves. The rate of buildup in this case will be approximately the reciprocal of the acoustic frequency. For frequencies near 10^{10} c.p.s., one sees that considerable buildup can be achieved within a pulse.

So far, we seem to have been limited by the phase matching condition to frequencies below about 10^{12} c.p.s. It is possible to go higher in

frequency by, for example, the Raman effect, which can be viewed as Brillouin scattering from optical phonons. This is possible because of the anomalous dispersion suffered by the Raman wave, which leads to \vec{k}_s taking on a wide range of values around resonance. Hence the condition $\vec{k}_0 = \vec{k}_{-1} + \vec{k}_s$ can be satisfied at very high frequencies. Any region of strong anomalous sound dispersion is likely to allow the phase matching condition, so that in such a region this type of interaction can generate acoustic frequencies considerably higher than those allowed by normal Brillouin scattering.

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